

This Page Is Inserted by IFW Operations
and is not a part of the Official Record

BEST AVAILABLE IMAGES

Defective images within this document are accurate representations of the original documents submitted by the applicant.

Defects in the images may include (but are not limited to):

- BLACK BORDERS
- TEXT CUT OFF AT TOP, BOTTOM OR SIDES
- FADED TEXT
- ILLEGIBLE TEXT
- SKEWED/SLANTED IMAGES
- COLORED PHOTOS
- BLACK OR VERY BLACK AND WHITE DARK PHOTOS
- GRAY SCALE DOCUMENTS

IMAGES ARE BEST AVAILABLE COPY.

**As rescanning documents *will not* correct images,
please do not report the images to the
Image Problem Mailbox.**

THIS PAGE BLANK (USPTO)

PATENT SPECIFICATION

774,805



Date of Application and filing Complete Specification June 24, 1954.

No. 18538/54.

Application made in United States of America on June 26, 1953.

Application made in United States of America on July 20, 1953.

Complete Specification Published May 15, 1957.

Index at

acceptance:—Classes 2(5), R1C(4: 5: 6: 8), R1T1, R2C(4: 5: 6: 8), R2T1, R6C(4: 5: 6: 8), R6T1, R29C(4: 5: 6: 8), R29T1, R30T1; 2(6), P4C(3: 4A: 4B: 8A: 8B: 8C: 10: 12X: 13A: 20A), P4D(2: 3A: 3B1: 3B3: 8), P4K9, P4P1(C: E1: X), P4P(4C: 5), P4S2, P9C(3: 4A: 4B: 8A: 8B: 8C: 10: 12X: 13A: 20A), P9D(1B1: 1B2: 1B3: 1X: 3: 8), P9K(4: 5: 6: 7: 8: 9: 11), P9P1(C: E1: X), P9P(4C: 5), P9S2; and 87(2), A1G(4X: 10), A1R14C(1A: 1X: 2), A2(B1: L).

International Classification:—B29f, g. C08f, g.

COMPLETE SPECIFICATION

Improvements in or relating to a Process for Producing a Thermosetting Resinous Molding Composition

We, AMERICAN CYANAMID COMPANY, a Corporation organised under the laws of the State of Maine, United States of America, of 30, Rockefeller Plaza, New York, State of New York, United States of America, do hereby declare the invention, for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:—

This invention relates to the production of thermosetting resinous molding compositions.

In the prior art preparations of resinous molding compositions, it is usually necessary to operate portions of the process batch-wise. Obviously, when it is required to carry out various steps of a procedure in a batch operation, an increase in production time and added costs are realized. Many pieces of equipment were also required to carry out the production of the molding composition.

When it was desired to produce a thermosetting molding composition from a dry resinous material, the starting material used was obtained by any process well known in the art, as, for example, spray-dried resin, pan-dried resin, etc. The dry finely ground resinous material was introduced with a filler, lubricants, catalysts, and the like, into a blending machine out of which was obtained a homogenized mixture. If desired, powdered, modifying resins could also be introduced into the mixing and blending machine. When the material was removed from the blending machine, it was introduced into such equipment as Banburys, heated differential rolls and the like so that a densification and partial solidification occurred. The densified material was then cooled and ground to obtain granulated particles. When a colored material was

desired, pigments and/or dyes were introduced with the dry resin, filler, and other additives in the initial blender.

An object of the present invention is to provide an improved procedure for the continuous densification of thermo-setting resinous material starting from either dry resins or resin syrups.

According to the invention, a process is provided for producing a thermosetting resinous molding composition, which comprises pulverizing thermosetting resinous material to a particle size within the range of 44 to 1410 microns, continuously introducing the pulverized material into a zone divided into a plurality of sub-heating zones maintained at different temperatures all of said temperatures being above 20° C. but below 150° C., continuous densification occurring in said zone, continuously removing the densified material from the zone in an extruded form and continuously cutting into pellets and cooling said material to room temperature.

The thermosetting resinous material which is pulverized may comprise a dry resin, a lubricant, and if desired, a filler. Alternatively, the thermosetting material may comprise a resinous syrup, a lubricant and if desired a filler, which are homogeneously blended at a temperature of 75° F. to 200° F., and continuously dried at a dry-bulb temperature of 100° F. to 215° F. and a wet-bulb temperature of 80° F. to 140° F., to a total volatile content not in excess of 15% by weight, prior to pulverization thereof.

It will be apparent that a colored molding composition may be obtained by adding dyes and/or pigments with the filler and other additives to the resinous material. During the extrusion process the resinous composition is

[Pric.

further blended and densified so that no additional processing steps, other than cutting into pellets and cooling of the material, are necessary in order to obtain a molding composition.

- 5 By extruding the material the necessity for regrinding, repulverizing and reblending as required in the prior art is obviated. The material removed from the extruded is in condition for molding when the extrusion is carried
10 out in such a way as to obtain pellets of the composition. If it is desired, fines, i.e., finely powdered resinous material of like composition may be added to the composition removed in pellet form from the extruded. However,
15 in order to obtain a suitable thermosetting resinous composition, it is not necessary to add the fines to the extruded material. By practicing our invention, a rapid and continuous densification of a thermosetting material is obtained. The necessity for the many large and costly machines previously required to produce a suitable molding composition has also been removed.

- The thermosetting resinous materials that
25 may be employed in the process of the invention are such as the phenol-aldehyde resins, aminotriazine-aldehyde resins, e.g., melamine-formaldehyde resin, aminotriazole-aldehyde resins, urea-aldehyde resins, urea-aminotriazine-aldehyde resins, aminodiazine-aldehyde resins, protein-aldehyde resins, e.g., casein-formaldehyde resins, resinous condensation products of aldehydes such as formaldehyde with polyamides as, for instance, malonic
35 diamide, maleic diamide, fumaric diamide or itaconic diamide. Examples of amino or amido compounds (amidogen compounds) that may be condensed with aldehydes such as hereinbefore mentioned
40 by way of illustration in forming a thermosetting resin, more particularly an aminoplast, are thiourea, diurea, diethylene triurea, methyl urea, acetyl urea, benzoyl urea, phenyl thiourea, asymmetrical diethyl urea, allyl urea, 2-chloro-
45 allyl urea, ethylidene urea, guanyl urea, biguanidine, aminoguanidine, melamine, triurea-ido melamine, ammeline, ammelide, melam, melam, melon, aminotriazoles or aminodiazines. Suitable mixtures of such
50 compounds also may be used. Phenol itself and various substituted phenols, for example the cresols or the xylenols may be condensed with aldehydes, e.g., formaldehyde or furfural to form thermosetting resins of the phenoplast
55 type which also find use in the process of the invention.

- Additional illustrative examples of thermosetting resinous compositions that may be used in the process of our invention are some
60 of the natural resins, e.g., shellac; the polymerizable unsaturated alkyd resins, e.g. ethylene glycol maleate, diethyl glycol fumarate, propylene glycol maleate phthalate, diethylene glycol fumarate adipate, propylene
65 glycol tetrahydroabietyl fumarate, butylene

glycol itaconate, butylene glycol tetra-hydroabietyl fumarate, ethylene glycol citraconate, diethylene glycol mesaconate, glycerol itaconate or ethylene glycol fumarate succinate; mixtures of such alkyd resins with others monomeric materials which are copolymerizable therewith, more particularly monomeric materials containing a $\text{CH}_2=\text{C}<$ grouping, e.g., an allyl grouping; and other thermosetting materials. Illustrative examples of monomeric materials containing $\text{CH}_2=\text{C}<$ grouping which may be copolymerized with a compatible unsaturated alkyd resin are styrent, dichlorostyrene, dimethylstyrene, triallyl phosphate, diallyl adipate, diallyl sebacate, diallyl citraconate, vinyl naphthalene, vinyl furane, ethyl vinyl ether, vinyl acetate, vinyl propionate, diallyl ether, methyl vinyl ketone, methylene malonic esters (e.g., methylene methyl malonate), acrylic and methacrylic esters (e.g., methyl, ethyl, propyl or butyl acrylates and methacrylates), acrylonitrile and acrylamide.

The dry thermosetting resins or resinous syrups employed in the process of the invention may be used by themselves or they may be used as mixtures of various thermosetting resinous material. In certain instances a molding composition may be desired produced from a single thermosetting resinous material. In other instances it may be desired to employ a combination of dry resinous materials to obtain a molding composition having certain desired properties. Accordingly, modified as well as unmodified resins find employment in our invention.

The amount of the thermosetting resinous material that may be used in the process of the invention may vary over very wide ranges. For example, if it is desired to produce a clear unfilled resinous material, it is obvious that 100% of the resin will be used. As used herein, percent indicates percent by weight unless otherwise noted. The amount of the filler that is added to the resinous material will depend upon the use to which the resin will be applied. Thus, when a material is desired that possesses high impact strength, a different amount and type of filler will be used than when a resinous material is desired that possesses other qualities, e.g., electrical properties. Thus, the amount of filler that is added may be as high as 80% by weight. The amount of the filler is, therefore, simply that proportion that causes the composition to possess the desired physical and/or other properties.

Illustrative examples of fillers that may be incorporated into the molding composition are alpha cellulose, wood flour, walnut shell flour, asbestos in the form of a powder or short or long fibers, finely divided silicon carbide, carbon black, diatomaceous earth, slate dust, powdered rutile, powdered or flaked mica, powdered quartz, cloth cuttings (e.g., cuttings of silk, rayon, wool, linen, cotton, nylon, or of

cloth made from glass fibres or from polymeric or copolymeric acrylonitrile fibers), ground cork and sand. As previously stated, depending upon the particular filler employed and the intended use of the molded product, the filler may constitute, for example, from 0% up to 80% by weight of the molding composition. It will also be noted that both organic and inorganic fillers may be utilized in the process of our invention.

Dyes, pigments, mold lubricants, and curing catalysts may also be introduced in the process of our invention with the resin and the filler in the pulverizing and blending operation.

Suitable curing catalysts that may be introduced into the reaction mixture are such as phthalic anhydride, ammonium chloride, ammonium silicofluoride and ammonium borofluoride. While the desired property of the molding composition will dictate the amount of catalyst introduced into the material, we have found that from about 0.01% to about 10% by weight of the catalyst will cover most eventualities. However, it is preferred that from about 1% to about 3% by weight of the catalyst be present in the resinous thermosetting material. Suitable mold lubricants are such as zinc stearate, glycerol monostearate, ceresin, carnauba wax, montan wax, calcium stearate and zinc palmitate. The lubricant as its name implies is present in the molding composition to facilitate the extrusion process and subsequent conversion into molded parts. From about 0.01% to about 5% by weight of the lubricant is generally sufficient amount. The preferred amount of lubricant is from about 0.3% to about 2.0% by weight. By carefully selecting the pigments, metallic powders and/or dyes introduced into the mixture, a final product of any desired color may be obtained. Illustrative examples of colorants that may be introduced into the mixture are such as cadmium yellow, cadmium red, cadmium maroon, titanium oxide, black iron oxide, chrome green, gold, silver, aluminum and copper. Suitable dyes are such as alizarine red, prussian blue, auramin, naphthol, malachite green. The amount of the colorants, catalysts and mold lubricants that are introduced into the mixture are such that the desired properties of the molded objects are obtained. Thus, for example, where a fast-curing object is desired, it is apparent that a larger amount of catalyst will be introduced into the mixture. The amount of the colorant may also vary depending upon the depth and shades of color desired. When a dark rich color is desired, colorants in an amount up to about 20% by weight may be used. Generally from about 0.5% to about 15% by weight of the colorants may be used to give any color desired. Obviously, when a non-colored molding composition is desired, no colorants will be introduced into the reaction mixture.

In carrying out the invention wherein a dry

thermosetting resin is used, the dry resinous material in combination with the mold lubricant and catalysts are introduced into a continuous pulverizer.

In carrying out the invention using a resinous syrup, the same is introduced into a suitable mixing chamber with a filler. There may also be introduced into the mixer catalysts, lubricants, and colorants. The temperature that is maintained in the reaction vessel may vary from 75° F. to 200° F. The material is maintained therein for a long enough period to insure a homogeneous mixture. If desired, pressure may be used during the blending of the syrup to insure thorough impregnation of the filler with the syrup. Of course, when pressure is applied to the vessel, elevated temperatures may be used. When a pressure is not applied to the vessel, it is preferred that the temperature therein be maintained within the range of from 100° F. to 160° F. After the material in the mixer has been thoroughly blended, it is then introduced into a continuous dryer wherein the volatile content is reduced to not in excess of 15% by weight. The volatile content of the material that is introduced into the continuous dryer is usually within the range of from 20% to 45% by weight. It is preferred, however, that the volatile content of the material introduced into the continuous dryer from the mixing be within the range of from 39% to 40% by weight. The continuous dryer is maintained at a temperature of from 100° F. to 215° F. dry-bulb, and from 80° F. to 140° F. wet-bulb, respectively. The period of time that the material remains in the continuous dryer is governed by the desired volatile content of the material removed therefrom. While the material may have as much as 15% by weight of volatiles, it is preferred that the volatile content of the material removed from the continuous dryer be not in excess of 8% by weight. In order to insure optimum drying in the continuous dryer, it is preferred that the temperature therein be maintained within the range of from 150° F. to 200° F. dry-bulb, and from 90° F. to 120° F. wet-bulb, respectively.

The original dry composition or the syrup composition after drying, as set forth above is then continuously run through a pulverizer so that a powder having a particle size within the micron range from 44 to 1410 is obtained. It is preferred that the size be in the micron range of from 44 to 149. Various alternative procedural steps may be employed in the process of our invention. For example, it is not necessary to introduce the catalyst and lubricant into the mixer along with the resin and filler. If desired, the lubricant and catalyst may be introduced into the material just prior to pulverizing. It is readily apparent that the colorant may also be introduced into the material at this stage of the process. When

a colorless molding composition is desired, colorants will not be added to the mixture.

The material, as it exists from the pulverizer, may be taken directly to the extruder. The extruded material is differentially heated so that there are a plurality of sub-heating zones. The temperature in the sub-zones may be controlled by the circulation therein of a heat exchange medium and the temperature of the zones may be controlled from 20° C. to 150° C. The temperatures are not necessarily fixed in the sub-zones of the extruder and in some instances increase in the direction of motion of the material, at other times decrease in the direction of motion of the material, and at other times may increase, decrease and then again increase in the direction of motion of the material. The temperature in the zones may vary in this manner according to the materials treated, the temperatures of the respective zones and other operating features required. Since the examples set forth hereinafter employ an extruder containing six zones, the temperatures employed in the various zones can be controlled either singly or two or more together. Thus in some of the examples only two temperature ranges are cited for the sub-zones. In these examples the adjacent zones are heated to the same temperature, and thus the temperature is controlled together. In other examples several temperatures for the sub-zones are employed and in these examples the temperatures of the zones are controlled singly. The temperature maintained in each of these heating zones of the extruder will vary depending upon the particular resinous material being extruded and also upon whether or not fillers and other additives are present. For optimum densification of the material, it is preferred that the sub-heating zones of the extruder be maintained at temperatures of approximately 20° C. to 30° C., from 60° C. to 135° C. and from 45° C. to 125° C., progressively. The length of time that the composition is maintained in the extruder depends upon the number of features. For instance, the revolutions per minute of the screws of the extruder, the pitch of the screw, the die through which the material is being extruded, the composition of the material being extruded, and the like. Of course, the material cannot remain in the extruder for such a period of time that it will set up and freeze the processing screws. The densified material may be extruded with or without a die. In the case where a die is not used, the densified extruded ribbon can be continuously cut in a cutter suitable for reduction to particles which can be subsequently continuously cooled. In the instance where a die is used, the densified material can be extruded in a variety of forms, for example, continuous rods or ribbons. When a die is used that contains a plurality of holes so that

rod-like extrusions are obtained, rotating knife blades attached to the end of the extruder cut the extruded material so that pellets of the molding composition are obtained. By controlling the die hole diameter, the revolutions per minute of the cutter, the number of blades on the cutter and the speed with which the material is being extruded, pellets of a predetermined size may be obtained. We have found that the surface appearance of a molded part will depend to a certain extent upon the length and diameter of the extruded pellets. Pellets having a diameter and length ranging from about 0.025 inches to about 0.10 inches are optimum for molding. Inasmuch as the material that is removed from the extruder is at an elevated temperature, the material may be cooled either before or after cutting into pellets and before it is packaged. A conventional method such as a water-cooled rotating cooler may be utilized to continuously cool the extruded material.

The plasticity of the extruded material is a function of (1), the formulation of the resinous composition, (2), the design of the die and the screws of the extruder, (3), the temperature of the heating zones, and (4), the revolutions per minute of the screws of the extruder. The plasticity of a material is that property which determines its rate of deformation under standard molding conditions.

In order to obtain the desired plasticity of the extruded material, various methods may be employed. For example, the speed of rotation of the screws may be increased or decreased if necessary, and also the design of the screw may be changed. Other methods of controlling the plasticity are by changing the die through which the material is extruded. The ratio of filler to resin may also be altered as well as the amount of mold lubricant added to control the plasticity of the material.

If desired, a vacuum may be applied to a section of the extruder in an amount sufficient to remove any volatiles and/or moisture released from the material during its extrusion. The pressure may vary from atmospheric pressure to an absolute pressure as low as 5 mm. of Hg. While the vacuum need not be applied during the extrusion process, it is preferred that this vacuum be present in order to obtain the optimum final bulk density of the extruded material.

In order that the invention may be fully understood, it will now be described with reference to the following examples, in which Ex. 1—4 and 7—9 start with dry resins, and Ex. 5—6 with resin syrups. All parts are by weight. The particular extruder employed in the following examples contains six sub-zones, but when two or more sub-zones adjacent to each other are maintained at the same temperature they are then considered as one sub-zone. It is impossible to determine the residence time of the material in each of the

sub-zones but the rate of total production per hour from the extruder is given in each example.

EXAMPLE 1

216 parts of dry powdered melamine-formaldehyde resin, 144 parts of dry powdered ether modified aniline resin (an aniline-formaldehyde condensation product modified with mono-*m-p*-cresyl glyceryl ether), 240 parts of alpha cellulose, 6 parts of calcium stearate, 6 parts of magnesium oxide and 1.2 parts of cadmium red were homogeneously ground and blended to a particle size within the range of from 44 microns to about 1410 microns. The material was then introduced into an extruder. Three sub-zones were employed and heated in the direction of motion of the material at 20° C., 100° C. and 70° C. When the statement is made that a zone is heated to 20° C. no circulation of a heating

material in this sub-zone is present and therefore the zone is maintained at approximately room temperature. The extruder screws were maintained at 60 rpm and the densified material was extruded at 120 parts per hour through a 2-inch die area containing 199 holes, each having a 0.072 inches diameter. An absolute vacuum of 455 mm. of Hg. was applied to a section of the extruder. Pellets of the molding composition having a diameter and length of about 0.072 inches were obtained by means of rotating knife blades attached to the end of extruder and continuously cooled.

In order to show the comparative properties of the continuously densified material prepared, the following table is set forth showing the properties of a like molding composition prepared in accordance with the prior art processes:—

	Prior art process	Continuously densified
Apparent density - - - - -	50—52	55
Molded appearance - - - - -	Fair	Fair
Minimum cure—minutes ($4 \times \frac{1}{8}$ " disc) - - - - -	2—2½	2½
Mold fouling - - - - -	Poor	Good
Insert cracking - - - - -	Poor	Fair

EXAMPLE 2

220 parts of a dry powdered melamine-formaldehyde resin, 47.5 parts of a dry powdered aniline-cresol resin (the condensation product of aniline, cresol and formaldehyde), 47.5 parts of a dry powdered aniline-shellac resin (the condensation product of aniline and formaldehyde modified with shellac), 96.5 parts of cotton flock, 288 parts of calcined asbestos and 7 parts of zinc stearate were homogeneously ground and blended as set forth in Example 1 to a particle size within the range of from 44 microns to about 1410 microns. The mixture was then continuously densified in an extruder. The sub-zones were heated in the direction of

motion of the material at 20° C., 120° C., 45° C. and 62° C. and under an absolute vacuum of 130 mm. of Hg. applied to a section thereof. The die through which the continuously densified material was extruded was the same as in Example 1 as was the particle size of the molding composition obtained. The pellets were continuously cooled upon removal from the extruder. The rate of production was 120 parts per hour.

The comparative properties of the resinous molding composition obtained with those of a like molding composition processed in accordance with prior art methods are set forth below:—

	Prior art process	Continuously densified
Apparent density - - - - -	54—59	70
Molded appearance - - - - -	Fair	Fair—Good
Minimum cure—minutes ($4 \times \frac{1}{8}$ " disc) - - - - -	2—2½	2
Mold fouling - - - - -	Poor	Good
Insert cracking - - - - -	Normal	Normal
Flexural strength - - - - -	6000—8000	7900—7951
Water absorption—% - - - - -	0.50	0.47
Dielectric constant (60 cycles) - - - - -	8.7	8.43

EXAMPLE 3

480 parts of a dry powdered melamine-formaldehyde resin, 120 parts of a dry powdered aniline-cresol resin, 400 parts of

wood flour, 1 part of calcium stearate, 0.5 part of cadmium red and 0.2 part of magnesium oxide were homogeneously ground and blended to a particle size of from 44 to 1410 microns

as set forth in Example 1. The mixture was then continuously densified in an extruder. The temperature of the sub-zones in the direction of motion was 100° C. and 65° C. No vacuum was applied to the extruder during the continuous densification. Pellets of the size obtained in Example 1 at the rate of 148 parts

per hour were removed from the extruder and continuously cooled.

Comparative properties of the continuously densified resinous molding composition with a like composition prepared in accordance with prior art methods as set forth below:—

15		Prior art process	Continuously densified
	Apparent density - - - - -	67—70	55
	Molded appearance - - - - -	Fair	Fair
	Minimum cure—minutes (4× $\frac{1}{8}$ " disc) -	2—2 $\frac{1}{4}$	2 $\frac{1}{2}$
	Mold fouling - - - - -	Poor	Good

20 EXAMPLE 4

The homogeneously ground and blended mixture obtained in Example 3 was introduced into the extruder wherein continuous densification occurred. In this instance the temperature of the sub-zones in the direction of motion of the material was 20° C., 100° C., 60° C. and the rate of production was 150 parts per hour. An absolute vacuum of 50 mm. of Hg. was applied to a section of the extruder. The continuously densified pellets removed from the extruder were then continuously cooled.

35 EXAMPLE 5

149.6 parts of a urea-formaldehyde resinous syrup, 50.4 parts of wood flour, 18 parts of barium sulfate and 1 part of zinc palmitate were introduced into a vessel maintained at a temperature of about 120° F.—130° F. wherein homogeneous blending occurred. The homogeneous mixture containing 40% by weight of volatiles was dried in a continuous dryer maintained at a dry-bulb temperature of 160° F. and a wet-bulb temperature of 115° F. The volatile content of the composition

removed from the dryer was less than 8% by weight. The material was then pulverized to a particle size of from 44—200 microns and was continuously introduced into an extruder. The temperature of the sub-zones in the direction of motion of the materials was 20° C., 85° C., and 80° C. and an absolute pressure of 25 mm. of Hg. was applied to a section of the extruder. The screws of the extruder were run at a temperature of 40 r.p.m. and the densified molding composition was continuously extruded at the rate of 125 parts per hour through a 2-inch die area containing 300 holes, each having a diameter of 0.072 inches. As the densified material was continuously extruded, it was sized by a rotating knife blade so that pellets having a diameter and length of about 0.072 inches were obtained which were then continuously cooled to room temperature.

The comparative properties of the continuously densified material prepared with those of a like resinous composition prepared in accordance with prior art methods is shown in the table set forth below:—

70		Prior art process	Continuously densified
	Apparent density - - - - -	58—66	61
	Molded appearance - - - - -	Fair	Fair
	Tons to close - - - - -	14+	14+
	Flow - - - - -	.030—.034	.033

75 EXAMPLE 6

1815 parts of a urea-formaldehyde resinous syrup, 389 parts of alpha cellulose, 12.8 parts of a 10% tetrachlorophthalic anhydride, 10.4 parts of hexamethylene tetramine, 6 parts of zinc palmitate and 131 parts of barium sulfate were introduced into a vessel maintained at a temperature of about 125° F. and was homogeneously blended therein. The homogeneous mixture containing 39% volatiles by weight was then dried in a continuous dryer maintained at a dry-bulb temperature of 160° F. and a wet-bulb temperature of 115° F. to a vola-

tile content of less than 8% by weight. After pulverizing the dried material to a particle size within the range of from 44 to 1410 microns, it was introduced into an extruder wherein continuous densification occurred. The temperature of the sub-zones in the direction of motion of the materials was 20° C., 120° C., 65° C. and 105° C. and the screws of the extruder were run at a speed of 60 r.p.m. The continuously densified material was extruded through a 2-inch die area containing 199 holes, each having a diameter of 0.072 inches. As the densified material was extruded, it was con-

tinuously cut into pellets and cooled to room temperature. Pellets having a length and diameter of about 0.072 inches were obtained.

EXAMPLE 7

The resin formulation of Example 1 was again employed and was continuously densified and extruded in the extruder employed in Example 1 under the same operating conditions except that the sub-heating zones of the extruder were maintained at temperatures of from 150° C. to 20° C., i.e. the first sub-zone of the extruder was heated to 150° C. and the last sub-zone of the extruder was not heated but was maintained at room temperature of approximately 20° C. The intermediate zones were all maintained at 100° C. An acceptable molding composition was obtained at the rate of 120 parts per hour.

EXAMPLE 8

28 parts of an ethylene glycol-maleic anhydride polyester resin obtained by heating equal molar proportions of the glycol and anhydride at elevated temperatures until an acid number of 37 was obtained, 2 parts of diallyl phthalate, 1 part of a urea-dicyandiamide formaldehyde resin, 19 parts of cotton flock and 50 parts of clay were homogeneously ground and blended to a particle size within the range of from 44 microns to 1410 microns. 2 parts of benzoyl peroxide, 0.2 parts of ditertiary butyl paracresol and 1 part of zinc stearate were blended into the resin formulation. The material was then introduced into an extruder having sub-zones maintained at temperatures of from 20° C. to 80° C. The extruder contained five separately

heat controlled sub-zones maintained from the feed end to the exit end of the extruder at the following respective temperatures: 20° C. i.e., no heating to the zone was supplied, 70° C., 65° C., 80° C. and 60° C. The material was extruded through a 2-inch die area containing 104 holes, each having a 0.122—0.144 inch tapered diameter. Pellets of the molding composition having a diameter and length of about 0.130 inches were obtained by means of rotating knife blades attached to the end of the extruder at a rate of about 118 parts per hour.

EXAMPLE 9

53.4 parts of a phenol-formaldehyde resin obtained by heating at reflux until two phase separation occurs, followed by removal of water and volatile materials by heating to 220° C. of 1 mol of phenol and 0.83 mols of formaldehyde in the presence of 1 part by weight of oxalic acid, 4.46 parts of hexamethylene tetramine, 42 parts of wood flour and 1 part of zinc stearate were homogeneously ground and blended to a particle size within the range of from 44 microns to 1410 microns. The material was continuously densified and extruded in the same equipment employed in the treatment of the polyester resinous composition except that the extruder sub-zones were maintained at the following temperatures: 20° C., 70° C., 95° C., 110° C. and 90° C. The resin was treated at a rate of 120 parts per hour.

The physical properties of the continuously densified resinous molding composition compared to those of a like material produced by prior art methods are shown in the following table:—

	Prior art process	Continuously densified
Apparent density - - - - -	64	35
Flexibility - - - - -	Poor	Fair
30-minute boil test:		
Cure time—minutes - - - - -	0.5	0.5
% H ₂ O absorption - - - - -	5.0	5.7
surface appearance - - - - -	OK	OK
Cure time—minutes - - - - -	1	1
% H ₂ O absorption - - - - -	4.3	4.7
surface appearance - - - - -	OK	OK
Cure time—minutes - - - - -	2	2
% H ₂ O absorption - - - - -	3.6	3.8
surface appearance - - - - -	OK	OK
Cure time—minutes - - - - -	3	3
% H ₂ O absorption - - - - -	3.4	3.4
surface appearance - - - - -	OK	OK

What we claim is:—

1. A process for producing a thermosetting resinous molding composition, which comprises pulverizing, thermosetting resinous material to a particle size within the range of 44 to 1410 microns, continuously introducing

the pulverized material into a zone divided into a plurality of sub-heating zones maintained at different temperatures all of said temperatures being above 20° C. but below 150° C. continuous densification occurring in said zone, continuously removing the densified material

- from the zone in an extruded form and continuously cutting into pellets and cooling said material to room temperature.
2. A process according to Claim 1, in which
- 5 the densified material is extruded as a continuous ribbon or rods.
3. A process according to Claims 1 or 2, in which a vacuum is applied to a section of said zone.
- 10 4. A process according to any of Claims 1 to 3, in which the sub-heating zones are maintained at temperatures of from 20° C. to 30° C., from 60° C. to 135° C. and from 40° C. to 125° C., progressively.
- 15 5. A process according to any of Claims 1 to 4, in which the thermosetting resinous material comprises a dry resin, a lubricant, and if desired a filler.
6. A process according to any of Claims 1 to
- 4, in which the thermosetting resinous material 20 comprises a resinous syrup, a lubricant and if desired a filler which are homogeneously blended at a temperature of 75° F. to 200° F., and continuously dried at a dry-bulb temperature of 100° F. to 215° F. and a wet-bulb 25 temperature of 80° F. to 140° F., to a total volatile content not in excess of 15% by weight, prior to pulverization thereof.
7. A process of producing a thermosetting resinous molding composition substantially as 30 hereinbefore described.
8. Thermosetting resinous molding compositions whenever produced by the process according to any of the preceding claims.

STEVENS, LANGNER, PARRY &
ROLLINSON,
Chartered Patent Agents,
Agents for the Applicants.

Leamington Spa: Printed for Her Majesty's Stationery Office, by the Courier Press,—1957.
Published at the Patent Office, 25, Southampton Buildings, London, W.C.2, from which
copies may be obtained.